Aerosol measurements in the IR: from limb to nadir?

A. Eldering, F. W. Irion, F. P. Mills, H. M. Steele, and M. R. Gunson

California Institute of Technology, Jet Propulsion Laboratory, MS 183-601 Pasadena , California 91109 Ph: (818) 354-4941 fax: (818) 354-5128 Annmarie.Eldering@jpl.nasa.gov

B. H. Kahn

UCLA Department of Atmospheric Sciences, 7127 Math Sciences, Los Angeles, California 90095

Abstract: Vertical profiles of aerosol concentration have been derived from the ATMOS solar occultation dataset. The EOS instrument TES has motivated studies of the feasibility of quantifying aerosols in nadir and limb emission measurements.

OCIS codes: (010.1100, 280.1100) Aerosol detection; (290.1090) Aerosol and cloud effects

1. Introduction

Knowledge of the composition and global distribution of aerosols is needed to improve our understanding of aerosol direct effects on Earth's radiation balance and indirect effects on cloud microphysics and precipitation [1]. Aerosols are also thought to play an important role in heterogeneous chemistry, effecting ozone concentrations [2]. More information about the global distribution and composition of aerosols is essential for making an assessment and prediction concerning the impact of human activities on a global basis. Remote sensing instruments are becoming widely discussed as a source of global aerosol information [3-5].

A methodology to retrieve vertical profiles of aerosol loading and composition from high spectral resolution infrared solar occultation measurements is presented and a study of the feasibility to retrieve aerosol characteristics from emission measurements is discussed.

The wavelength dependent aerosol extinction in the 800 to 1250 cm⁻¹ region has been derived from ATMOS high spectral resolution infrared transmission measurements. Using models of aerosol and cloud extinction, weighted nonlinear least squares fitting was performed to determine the aerosol volume columns and vertical profiles of stratospheric sulfate aerosol. Modeled extinction from an aerosol modeled as 75% H₂SO₄ acid shows excellent agreement with the measurements, and the derived aerosol volumes for 1992 measurements are consistent with other measurements after the eruption of Mt. Pinatubo.

Instruments that are currently under construction such as the Tropospheric Emission Spectrometer (TES) of NASA's Aura satellite will also be making measurement in the thermal infrared region with very high spectral resolution. Although the radiative transfer problem is more complex, there is some potential for aerosol characterization from these measurements. Sensitivity studies are being conducted and current results suggest that aerosol optical depths greater than 0.01 will be detectable.

2. ATMOS results

ATMOS is a high resolution interferometer that made measurements from 2.2 to $16~\mu m$ with a resolution of $0.01~cm^{-1}$. The ATMOS data set was collected from the Space Shuttle in four missions, with data collection lasting from 24 hours to 11 days in 1985, 1992, 1993, and 1994. Approximately 350 occultations, each containing 50 to 100 spectra, are available. During a single occultation, spectra are collected through one of six optical bandpass filters which are 600 to $1600~cm^{-1}$ wide. These measurements of transmission at high spectral resolution will be used to study aerosol characteristics.

The term continua spectra, K_{meas} , will refer to the frequency dependent spectra of continua absorption which is attributed to aerosol. The continua absorption is the residual after accounting for discrete line absorption by atmospheric gases (including absorption in the far wings of pressure broadened lines and pressure induced absorption by N_2 and O_2) and broadly absorbing gases such as CFCs.

The transmission of solar radiation at a frequency, v, through an atmosphere containing gases, j, with absorption coefficients, κ_j and number density of the gas, g_j , and aerosols of i types along optical slant path x, is given by:

$$T(v) = \exp[-\sum_{j} \{ \int_{x} \kappa_{j}(v, x) g_{j}(x) dx \}] * K_{meas}(v)$$
 (1)

We have developed a method to obtain the continua spectra from high spectral resolution transmission measurements. The spectrum is divided into 2 cm⁻¹ intervals that are treated individually. Trace gases within each window are retrieved independently and the continuum level and its associated error are determined for each interval across the spectrum. The errors in the continuum are a reflection of the ability to account for all gas absorption in the window and are determined by the covariance matrix in the fitting of the trace gases.

Figure 1 is an illustration of a continuum spectrum from 1992 and the best fit aerosol models. This measurement shows evidence of the increase in stratospheric sulfuric acid (SSA) after the eruption of Mt. Pinatubo in 1991. The slant column associated with the best fit is reported in Figure 1.

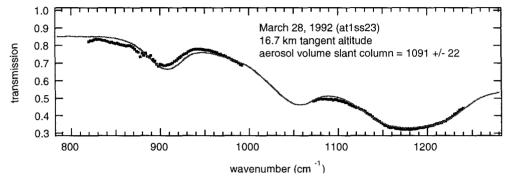


Fig. 1. Continuua spectra and best fit aerosol model of sulfuric acid (solid line) with a composition of 75% H_2SO_4 by weight.

An occultation measurement consists of a series of spectra that are taken during local sunrise or sunset, and the spectra are identified by the tangent height of the optical path above the Earth. In satellite occultation measurements, the geometric slant path distances, dx, are heavily weighted to altitudes near the tangent altitude and a single spectrum cannot be used to derive the vertical profile of $g_j(x)$ or $N_i(x)$. Using an assumed vertical profile, nonlinear least squares fitting is performed to find the slant column abundance from each spectra in an occultation. The set of slant columns and the matrix of slant path distances are inverted with a linear equation solver to yield a vertical profile of the aerosol concentration or gas volume mixing ratios. The fitting and inversion processes are then repeated using the retrieved vertical profile as the initial guess.

An illustration of a retrieved vertical profile of aerosol volume for an ATMOS measurement from 1992 is provided in Figure 2. The vertical profiles of SSA shown in Figure 2 shows a peak in the concentration of SSA of about $2.0 \, \mu m^3 \, cm^{-3}$) consistent with reports of the aerosol volume shortly after the eruption of Mt. Pinatubo and the decay of the aerosol layer in the following years [6].

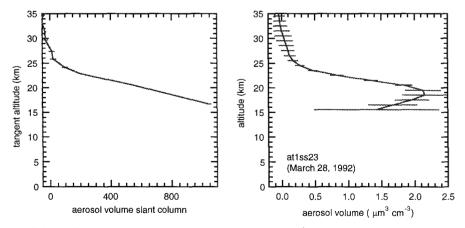


Fig. 2. Aerosol slant columns and vertical profile of stratospheric sulfuric acid aerosol from ATMOS measurements.

3. Tropospheric Emission Spectrometer (TES)

TES will acquire high resolution infrared spectra of the naturally occurring infrared emission from the Earth's atmosphere. The instrument is a Fourier Transform spectrometer that measures spectral radiance in the 650 - 3050 cm⁻¹ (3.3 - 15.4 µm) spectral range with a spectral resolution of 0.1 cm⁻¹ (nadir viewing) or 0.025 cm⁻¹ (limb viewing). Spectra with 0.1 cm⁻¹ and 0.025 cm⁻¹ resolution are acquired in 4 s and 16 s respectively. High spectral resolution and broad spectral coverage are essential for measuring the key atmospheric species over the 0 to 30 km altitude range that TES observes. It also allows a comprehensive survey of the entire suite of molecules found in the troposphere and lower stratosphere. The high spectral resolution minimizes detection interference between species and insures that high vertical resolution is maintained over the observed altitude range.

Although TES was developed with high spectral resolution so that vertical sounding of gases could be performed, the high spectral resolution is also favorable for the characterization of aerosol. If the gas signal can be accounted for accurately, spectrally dependent aerosol signatures can also be revealed. The radiative transfer for thermal emission measurements including scatterers is more complex that the transmission measurements of ATMOS, but adequate models are available.

Some analysis of nadir sounding data has been performed to study clouds [7,8] and stratospheric sulfuric acid after the eruption of Mt. Pinatibo [9]. These analyses rely on small sets of frequencies and use the ratio of emissivities or brightness temperatures to characterize the clouds or aerosols. We are proposing a method that would use more frequencies in the window region, allowing for greater discrimination between aerosol compositions. Calculations have been carried out to examine the sensitivity of nadir emission measurements to stratospheric sulfuric acid aerosol, tropospheric dust (see Figure 3), and cirrus clouds. These calculations show that the radiance differences in the window region of 800 to 1000 cm⁻¹ are on the order of 5x10⁻⁸ W cm⁻² sr⁻¹ cm for optical depths of 0.01. The spectral dependence of the radiance differences is material dependent. The sensitivity of TES and approaches for characterizing aerosols will be discussed.

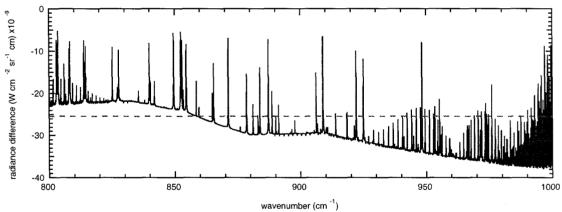


Fig. 3. Calculated radiance difference between a no aerosol case and a case with tropospheric dust with an optical depth of 0.01. The TES NESR is shown as a dotted line.

7. References

- 1. R.J. Charlson and J. Heintzenberg (Eds.), *Dahlem Workshop Report on Aerosol Forcing of Climate*, 416 pp. John Wiley, New York, (1995). 2. F.J. Dentener, G.R. Carmichael, Y. Zhang, J. Lelieveld, and P.J. Crutzen, "Role of mineral aerosol as a reactive surface in the global troposphere," J. Geophys. Res. **101**, 22869-22889 (1996).
- 3. Y.J. Kaufman, D. Tanré, H.R. Gordon, T. Nakajima, J. Lenoble, R. Frouin, H. Grassl, B. M. Herman, M. D. King, and P. M. Teillet, "Passive remote sensing of tropospheric aerosol and atmospheric correction for the aerosol effect," J. Geophys. Res. 102, 16815-16830 (1997).

 4. M. King, Y. Kaufman, D. Tanré, and T. Nakajima, "Remote sensing of tropospheric aerosols from space: past, present, and future," AMS Bulletin, 80, 2229-2259 (1999).
- 5. D.L Hartmann and P. Mouginis-Mark, "Volcanoes and climate effects of aerosol", in *EOS Science Plan*, Edited by M. King, (1999).
 6. A. Lambert, R.G. Grainger, C.D. Rodgers, F.W. Taylor, J.L. Mergenthaler, J.B. Kumer, and S. T. Massie, Global evolution of the Mt. Pinaubo volcanic aerosols observed by the infrared limb-sounding instruments CLAES and ISAMS on the Upper Atmosphere Research Satellite, J. Geophys. Res. 102, 1495-1512 (1997).
- 7. A. J. Baran, J. S. Foot, and David L. Mitchell, "Ice-crystal absorption: a comparison between theory and implications for remotesensing," Applied Optics, 37, 2207-2215 (1998).
- 8. C. J. Stubenrauch et al., "Retrieval of cirrus ice crystal sized from 8.3 and 11.1 μm emissivities determined by the improred initialization inversion of TIROS-N Operational Vertical Sounder observations," J. Geophys. Res. **104**, 31793-31808 (1999).
- 9. S. A. Ackerman, "Remote sensing aerosols using satellite infrared observations," J. Geophys. Res. 104, 17069-17079 (1997).